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Pd^{115} YIELD IN THERMAL NEUTRON FISSION OF U^{235}

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ABSTRACT

The formation of Pd^{115} in thermal neutron fission of U^{235} was studied quantitatively. A procedure was developed which rapidly separated palladium from daughter elements silver and cadmium. Separations were made from 2.8-120 seconds after fission. The growth of Cd^{115} from palladium separated at various times constituted a decay curve for Pd^{115}. The half-life thus determined was $40.5^{+4.0}_{-3.0}$ seconds, a value which agrees with that previously reported. The cumulative fractional yield of Pd^{115} compared to the total Cd^{115} yield was 90.5 ± 7.2 %. Estimation of the most probable nuclear charge (Z_p) must await information on the contribution of other members to the yield of the 115 mass chain.

SUMMARY

Problem

As part of a general study of nuclear charge distribution in the region of symmetric fission of uranium, information on the formation of a palladium isotope (mass 115) was required.

Findings

A rapid radiochemical separation method was developed to study the formation of this palladium isotope. The half-life of this fission product was confirmed and its cumulative contribution to the mass 115 chain yield was determined.

INTRODUCTION

The distribution of nuclear charge in near-symmetric thermal neutron fission of U^{235} is being studied. For this study, information is needed on the cumulative fission yield of Pd^{115}.

Alexander and co-workers[1] reported the formation of Pd^{115} upon bombardment of natural uranium with deuterons. Palladium was isolated free from silver and cadmium within about 3 minutes after fission. By the periodic extraction of silver decay products and purification and counting of 2.2-day Cd^{115} after decay of 21.1-minute Ag^{115}, a half-life of 45 ± 3 seconds was determined for Pd^{115}.

The present work was aimed at quantitatively examining the formation of Pd^{115} as quickly after fission as possible. Speed was essential to reduce the probability of failure to observe the contribution to the yield by any as yet unidentified short-lived isomer.

A rapid method was developed for the separation of palladium from its descendants, silver and cadmium. The procedure was based upon the reduction and simultaneous removal from solution of palladium by copper powder.[2] The reduction of silver was inhibited by complexation with bromide; the oxidation potential of cadmium is unfavorable for its reduction by copper. The unreduced elements remained essentially quantitatively in solution.

With this method the quantity of descendant Cd^{115} was measured in palladium samples isolated at various times after irradiation. The curve which resulted from a plot of Cd^{115} activity as a function of separation time constitutes a decay curve for Pd^{115}. From this data together with a separate measurement of the total Cd^{115} yield, the cumulative fractional yield of Pd^{115} was computed. The difficulty of using this cumulative fractional yield to estimate the most probable nuclear charge (Z_p) is described.

EXPERIMENTAL

Chemical Solutions

The weights given in the solutions listed below refer to the metallic element. All reagents used were of analytical grade.

Uranyl bromide was prepared from 93.17 % enriched U^{235} metal. The metal was dissolved in conc. HNO_3, and excess HNO_3 was removed by several evaporations with conc. HBr. The dried salt was finally dissolved in conc. HBr at a concentration of 400 mg/ml.

Ruthenium, rhodium and palladium carriers were prepared from the respective metals. After dissolution they were converted to chlorides and finally dissolved in 3 N HCl at concentrations of 10, 6.1 and 10.0 mg/ml, respectively.

Silver carrier was prepared from $AgNO_3$ in conc. HBr at a concentration of 10 mg/ml.

The conc. HBr-Br_2 solution contained 1 drop of Br_2/ml.

Procedure for Pd^{115} Yield Determination

A solution of enriched uranium (250 λ), of ruthenium, rhodium, silver, and palladium carriers (100, 150, 100 and 200 λ, respectively), and of conc. HBr-Br_2 (500 λ) was contained in a pneumatically-driven sample carrier (rabbit). Each rabbit was irradiated for 5 seconds in the Vallecitos Nuclear Test Reactor in a flux of $\sim 10^{12}$ neutrons cm^{-2} sec^{-1}. The cadmium ratio for gold was determined to be 2.7. A gold foil taped to the rabbit served as a monitor of the number of fissions in the sample as before.[3]

At the end of the irradiation the rabbit was transferred in about 1 second a distance of 50 feet to the laboratory. The irradiated solution was transferred by suction to a tube containing 10 ml of hot conc. HBr, and in most cases the rabbit was washed with 1 ml of conc. HBr. The combined solution was passed through 2 grams of copper powder in a filtration apparatus[2] at a definite time after the end of the irradiation. Passage of the solution through the copper was complete in less than 1 second. For separations which occurred more than several seconds after irradiation the copper bed was washed immediately with 5 ml of hot conc. HBr.

After separation the copper was transferred to a centrifuge tube which contained a definite volume of standardized cadmium carrier. Several hours after irradiation the copper was dissolved with 10 ml conc. HNO_3, evaporated to dryness and then brought twice more to dryness after the addition of 5-ml volumes of conc. HCl. The residue was dissolved in 125 ml of 4 N HCl, and both copper and palladium were quantitatively precipitated by bubbling H_2S gas for 10 minutes through the previously warmed solution. The insoluble sulfides were collected on sintered glass and saved for the palladium-yield determination.

The clear filtrate containing the cadmium was alkalinized with NaOH pellets. H_2S gas was further bubbled through the filtrate for several minutes, and the precipitated cadmium sulfide was separated by centrifugation. This precipitate was dissolved in 1 ml of conc. HCl and the solution was analyzed for Cd^{115} by the radiochemical procedure of Hicks.[4] The beta-ray activity was measured on a gas-flow proportional counter for 1 to 2 months and the decay curve was resolved into the 2.3- and 43-day isomers of Cd^{115}. The count rate of the shorter-lived isomer was corrected for decay from the time of irradiation, for cadmium carrier yield in the radiochemical procedure, and for palladium yield (see below), and was normalized to 10^{12} fissions.

The palladium carrier yield in the initial separation was determined spectrophotometrically[5] with corrections by the isotopic dilution technique for losses incurred in the purification process. The purification was performed as follows: The copper-palladium insoluble sulfide fraction was dissolved from the sintered glass filter with aqua regia after the addition of a known quantity of Pd^{109}. This solution was evaporated to dryness and the residue was dissolved in 20 ml of water.

Two ml of 1 % dimethylglyoxime in ethyl alcohol were added and the solution was extracted two times with 10-ml portions of chloroform. The chloroform extract was washed three times with 5 ml of 0.3 N HCl and the palladium was then extracted into 10 ml NH_4OH. After the ammonia extract was heated to a boil, H_2S gas was bubbled through it, and conc. HCl was added dropwise until the palladium sulfide precipitate appeared. After 3 minutes more of H_2S bubbling, the precipitate was separated from the solution. The sulfide was dissolved in 2 or 3 drops of aqua regia and carefully evaporated to complete dryness. The residue was dissolved and brought to a definite volume with 1.5×10^{-3} N HCl. Colorimetric[5] and Pd^{109} analysis of aliquots of this solution provided the values for the palladium yield determination.

Total Cd^{115} Yield Determination

Two rabbits with contents as above including the various carriers were irradiated for 40 seconds. Several hours after the irradiation

they were analyzed for Cd^{115}. The count rate was corrected for decay and cadmium carrier yield, and again was normalized to 10^{12} fissions.

RESULTS AND DISCUSSION

In the rapid separation procedure of palladium from silver and cadmium, 75-80 % of the palladium was reduced and retained by the copper bed. Preliminary experiments with radioactive tracers had indicated that contamination by cadmium and silver was 4.2×10^{-2} % and 0.95 %, respectively. Ruthenium and rhodium, potential palladium precursors, are associated with the copper bed to the extent of 3.0 % and 14.3 %. As discussed below, error from contamination by ruthenium and rhodium was not evident in the decay measurements of Pd^{115}. Half-lives of isotopes of these elements with a mass number of 115 are probably too short for them to be contaminants in this procedure.

The counting rate of Cd^{115} for 16 separations extending in time from 2.8-120 seconds after fission appears in Fig. 1. The time of separation was the time between the end of irradiation and filtration through the copper bed. The relationship between the logarithm of the counting rate and the separation time is linear. Analysis by the method of least squares gives a half-life of $40.5^{+4.0}_{-3.0}$ sec for Pd^{115} which agrees with the previously reported half-life.[1] No shorter-lived isomer of Pd^{115} was detectable, nor was interference from precursors evident.

The counting rate of Cd^{115} extrapolated to the end of the irradiation was corrected for the formation and decay of Pd^{115} during the course of the 5-second irradiation time. This corrected value was compared with the counting rate for the total Cd^{115} formed. The cumulative fractional chain yield of Pd^{115} thereby determined was 90.6 ± 7.2 %. Formerly, under the assumption that the distribution of nuclear charge is Gaussian and that the width parameter σ is 0.62, values of Z_p were computed from cumulative fractional yields.[6,7] However, recent information points to the unreliability of this approach; in the 131-136 mass chains the values of σ were determined to vary over the range of 0.28-0.69.* To illustrate the sensitivity of Z_p over this range of σ values, the above treatment gives values of Z_p which vary from 45.58-46.08.

*Strom, P. O., et al., to be published.

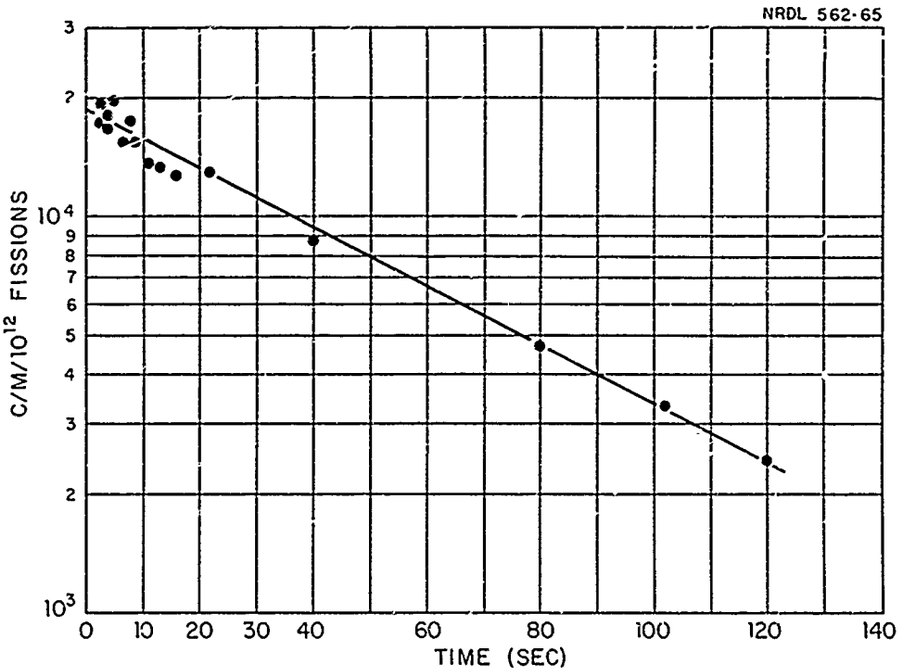

Fig. 1 The Growth of Cd^{115} From Pd^{115} at Various Times of Pd Separation

More adequate characterization of Z_p for mass 115 will depend upon a separate evaluation of the contribution of other chain members to the total yield. Currently experiments are underway to measure the independent yield of the two Ag^{115} isomers; subsequently, if practicable, the independent yield of Cd^{115} will be determined.

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